Terahertz optical gain based on intersubband transitions in optically-pumped semiconductor quantum wells: Coherent pump-probe interactions

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Abstract

Terahertz optical gain due to intersubband transitions in optically-pumped semiconductor quantum wells (QW's) is calculated nonperturbatively. We solve the pump-field-induced nonequilibrium distribution function for each subband of the QW system from a set of rate equations that include both intrasubband and intersubband relaxation processes. The gain arising from population inversion and stimulated Raman processes is calculated in a unified manner. We show that the coherent pump and signal wave interactions contribute significantly to the THz gain. Because of the optical Stark effect and pump-induced population redistribution, optical gain saturation at larger pump intensities is predicted.

1

In recent years there has been considerable interest in intersubband-transition-based infrared semiconductor quantum well (QW) lasers because of their potential applications. In the mid-infrared frequency range, semiconductor quantum cascade lasers based on unipolar electrical injection were demonstrated experimentally.[1-3] In these studies, optical gain is due to global k-space population inversion between the lasing subbands. In addition to the study of the electrically injected intersubband lasers, optically pumped intersubband lasers were also proposed[4,5] and experimentally demonstrated in the mid-infrared range.[6] As in the electrical pumping case, the appearance of the optical gain in the optically pumped QW system was mainly attributed to the pump-field-induced population inversion.[4-6] In a third-order perturbative theory, it was suggested that stimulated Raman scattering in QW systems can produce net optical gain, and the maximum gain of the QW system is linearly proportional to the pump intensity.[7,8] In such a nonlinear optical scheme, the appearance of optical gain that may lead to intersubband Raman lasers does not rely on the population inversion. Since, in the resonant Raman process (Raman gain is the largest in this case), the strong pump field induces a substantial population redistribution among subbands in the QW system, it seems that a realistic estimate of the optical gain has to include this effect. Perturbative calculations may overestimate the Raman gain. Also, the optical Stark effect was not included in the previous perturbative treatment.[7,8] That effect certainly influences the optical gain of the optically-pumped QW system.

In this paper we present a nonperturbative calculation of terahertz gain of optically-pumped semiconductor step quantum wells. The optical gain arising from pump-light-induced population inversion and stimulated Raman processes is calculated in a unified manner. Limiting optical transitions within the conduction band of the QW, we solve the pump-field-induced nonequilibrium distribution function for each subband of the QW system from the rate equations. Both intrasubband and intersubband relaxation processes in the quantum well system are included. Taking into account the coherent interactions between pump and THz (signal) waves, we calculate the susceptibility of the QW system for the THz field. We show that the coherent wave interactions (resonant stimulated Raman processes)

contribute significantly to the THz gain in addition to the contributions from the population inversion. Owing to the optical Stark effect and pump-light-induced population redistribution, the maximum gain is in general not linearly proportional to the pump intensity. When the pump field is sufficiently strong, gain saturation is predicted.

We consider an asymetric step QW structure with three subbands within the conduction band. The two upper subbands are the lasing states, and the subband energy separation (E_{32}) at zero wave vector of electrons $(k_\| = 0)$ lies in the THz frequency range (1-10 THz or 4-40 meV). The eigenenergy (E_m) and corresponding wave function (ψ_m) of the QW are calculated from the effective-mass Schrödinger equation coupled with Poisson equation. The exchange-correlation effect is included in the local density approximation.[9] The conduction band nonparabolicity is taken into account by using a subband-energy-dependent effective mass.[10] A strong pump field of frequency ω_p $(\hbar\omega_p \approx E_{31})$ drives the QW system (see the inset of Fig. 1). Assuming the electromagnetic coupling from the signal (THz) wave to the pump field is weak, we derive the rate equations for the electronic distribution functions $[f_m(k_\|) \equiv f_m(m = 1, 2, 3)]$ for the three subbands from the single-particle density matrix formalism.[11] Including both intrasubband and intersubband relaxation processes, the rate equations read as

$$\frac{\partial f_1}{\partial t} = -W_{12}(f_1 - f_2) - W_{13}(f_1 - f_3) - \frac{f_1 - f_1^{\rm F}}{T_{cc}} + \frac{f_2}{\tau_{21}} + \frac{f_3}{\tau_{31}} \,, \qquad (1)$$

$$\frac{\partial f_2}{\partial t} = W_{12}(f_1 - f_2) - \frac{f_2 - f_2^{\rm F}}{T_{cc}} - \frac{f_2}{\tau_{21}} + \frac{f_3}{\tau_{32}} \,, \qquad (2)$$

$$\frac{\partial f_3}{\partial t} = W_{13}(f_1 - f_3) - \frac{f_3 - f_3^{\rm F}}{T_{cc}} - \frac{f_3}{\tau_{31}} - \frac{f_3}{\tau_{32}} \,, \qquad (3)$$

where

$$W_{mn} = \frac{2\Gamma_{mn}}{\hbar} \frac{|H'_{mn}|^2}{[\hbar\omega_p - E_{mn}(k_\|)]^2 + \Gamma_{mn}^2} \qquad (4)$$

is the pumping rate from subband m to subband n. In the above equations T_{cc}, τ_{mn}, and Γ_{mn} are the intrasubband carrier-carrier scattering time, intersubband relaxation time, and

3

line broadening, respectively. In the electrical dipole approximation, the matrix element of the light-quantum-well interaction Hamiltonian is $H'_{mn} = -E_p\mu_{mn}$, where E_p denotes the amplitude of the pump electric field that is polarized perpendicular to the wells (in the z axis) [since the local-field effect is negligibly small for the electron densities used in this paper, E_p is taken to be the external field] and $\mu_{mn} = e\int \psi_m(z)z\psi_n(z)dz$ is the dipole moment between subband m and subband n. The quantities $f_m^F(m = 1,2,3)$ are considered to be the quasi-equilibrium distribution functions which are related to the Fermi-Dirac functions $f_m^{(0)}(m = 1,2,3)$ in the absence of pump field. Thus, for a given temperature and electron density, the steady-state nonequilibrium distribution function for each subband is uniquely determined from Eqs. (1)-(3). Here we would like to emphasize that in our treatment the total particle number in our QW system is conserved, i.e., $f_1 + f_2 + f_3 = f_1^{(0)} + f_2^{(0)} + f_3^{(0)}$.

Taking into account the pump and THz wave interactions in the QW system,[11] the off-diaganol element of the density matrix operator between subband 3 and subband 2 at the THz frequency ω is given by

$$\rho_{32}(\omega) = \frac{[f_2 - f_3 - \mathcal{R}(\omega,\omega_p)](-\mu_{32}E_\omega)}{\hbar\omega - E_{32}(k_\parallel) + i\Gamma_{23} - \Delta(\omega,\omega_p)} \equiv -A(\omega,\omega_p,k_\parallel)\mu_{32}E_\omega , \tag{5}$$

where E_ω is the THz field amplitude, and

$$\Delta(\omega,\omega_p) = \frac{|H'_{31}|^2}{\hbar(\omega_p - \omega) - E_{21}(k_\parallel) - i\Gamma_{12}} + \frac{|H'_{21}|^2}{\hbar(\omega_p + \omega) - E_{31}(k_\parallel) + i\Gamma_{13}} , \tag{6}$$

$$\mathcal{R}(\omega,\omega_p) = \frac{|H'_{31}|^2(f_1 - f_3)}{[\hbar(\omega_p - \omega) - E_{21}(k_\parallel) - i\Gamma_{12}][\hbar\omega_p - E_{31}(k_\parallel) - i\Gamma_{13}]}$$

$$+ \frac{|H'_{21}|^2(f_1 - f_2)}{[\hbar(\omega_p + \omega) - E_{31}(k_\parallel) + i\Gamma_{13}][\hbar\omega_p - E_{21}(k_\parallel) + i\Gamma_{12}]} . \tag{7}$$

$\Delta(\omega,\omega_p)$ in Eq. (5) is responsible for the optical Stark effect while $\mathcal{R}(\omega,\omega_p)$ gives rise to the Raman gain of the QW system. From Eq. (5) we define the susceptibility for the THz probe field as

$$\chi(\omega) = -\frac{2|\mu_{23}|^2}{\epsilon_0 L_w} \int \int \frac{d^2k_\parallel}{(2\pi)^2} A(\omega,\omega_p,k_\parallel) , \tag{8}$$

L_w being the width of the active layer. Therefore, the optical gain is given by

I

$$G(\omega) = -\frac{\omega}{c_0 n_b}\text{Im}[\chi(\omega)] .$$ (9)

Note that, if we neglect $\Delta(\omega,\omega_p)$ in Eq. (5) and let $f_m = f_m^{(0)}$ in Eqs. (5) and (7), the Raman gain given in Eq. (9) is linearly proportional to the pump intensity or $|E_p|^2$, as in Refs. [7] and [8]. Also we notice from Eq. (5) that the influence of the pump field to the optical gain (or loss) is through the combination of stimulated Raman processes, the optical Stark effect, and the population redistribution. Therefore, the optical gain is in general not linearly dependent on the pump intensity, as we will see in the following.

Using Eqs. (1)-(9) we calculated the THz gain spectrum of a GaAs/AlGaAs step QW at different pump frequencies and intensities. The QW structure used in our calculations is similar to that in Ref. [12]. The deep well width is 65 Å and the shallow well width is 130 Å. The barrier height (relative to the deep well) is 225 meV and the step barrier height is 140 meV. Using these parameters we found that at $k_\parallel=0$ $E_{32} \approx 25.0$ meV and $E_{31} \approx 124.9$ meV (the effective mass for each subband was also determined). The other parameters employed in our calculations are: $\Gamma_{12} = \Gamma_{13} = \Gamma_{23}=3.0$ meV, $T_{cc}=0.6$ ps, $\tau_{21}=1.0$ ps, $\tau_{31}=1.2$ ps, and $\tau_{32}=1.5$ ps.[6] To avoid the strong plasma absorption for the THz field, we used a small sheet electron density of 5.17×10^{10} cm^{-2}. The temperature was taken to be T=100 K. In Fig. 1 we show the THz gain spectra (solid curves) of our QW structure at the pump photon energy of $\hbar\omega_p = E_{31}$ for different pump intensities, namely, 0.1, 0.3, and 1.0 MW/cm^2. For comparison, we also plot in dashed curves the calculated THz gain without stimulated Raman contributions. It appears from Fig. 1 that varying the pump intensity leads to not only a change in the maximum value of the THz gain but also a notable shift of the peak position. The blueshift of the peak position in the gain spectrum is due to the optical Stark effect. We also note from Fig. 1 that, as the pump intensity increases from 0.1 to 1.0 MW/cm^2, the maximum gain first increases and then decreases. When the pump intensity is further increased, the gain decreases monotonically. This gain saturation stems from both optical Stark effect and pump-induced population redistribution. (Gain saturation due to pump field depletion is not included in our theory. This effect is important when

the conversion efficiency is large. We expect that this is not the case in our calculations.) In comparing solid and dashed curves in Fig. 1, it is evident that the stimulated Raman scattering contributes significantly to the THz gain.

To see more clearly the saturation behavior of the THz gain, we show in Fig. 2 the gain of our QW system as a function of the pump intensity for different probe frequencies, i.e., $\hbar\omega$ =20, 25 and 30 meV. The maximum gain versus the pump intensity is also displayed. In calculating Fig. 2 a pump photon energy of $\hbar\omega_p = E_{31}$ was used. We see from Fig. 2 that, when the pump field is very weak, the gain almost linearly increases with an increase in the pump intensity. This should be expected because in the weak pumping case the perturbation theory is valid. However, when the pump intensity is increased to certain values (say $\geq \sim 0.2$ MW/cm^2), our calculated results deviate substantially from those predicted from the perturbation calculations.

In conclusion, we have calculated terahertz gain of optically-pumped semiconductor step quantum wells. Both the coherent pump and probe wave interactions and pump-light-induced population redistribution are taken into account. Our calculations show that the optical gain is strongly dependent on the pump intensity and frequency, and the maximum gain is not linearly proportional to the pump intensity even when the pump field is moderately strong. Because of the coherent pump and THz wave interactions as well as the light-induced population redistribution among the subbands, gain saturation is predicted.

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FIGURES

FIG. 1. THz gain spectra of a GaAs/AlGaAs step QW with a subband separation of $E_{32} \approx 25.0$ meV at a pump photon energy of $\hbar\omega_p = E_{31}$ for different pump intensities, i.e., 0.1, 0.3, and 1.0 MW/cm^2. The dashed curves represent the results without the contribution from the stimulated Raman scattering. In the inset, a schematic diagram shows a three-subband step QW driven by a pump field and THz emission due to transitions from subband E_3 to subband E_2.

FIG. 2. THz gain spectra of a GaAs/AlGaAs step QW as a function of the pump intensity for different probe frequencies, i.e. $\hbar\omega = 20$, 25, and 30 meV. The pump photon energy is $\hbar\omega_p = E_{31}$. The dashed line shows the maximum gain of the QW system.

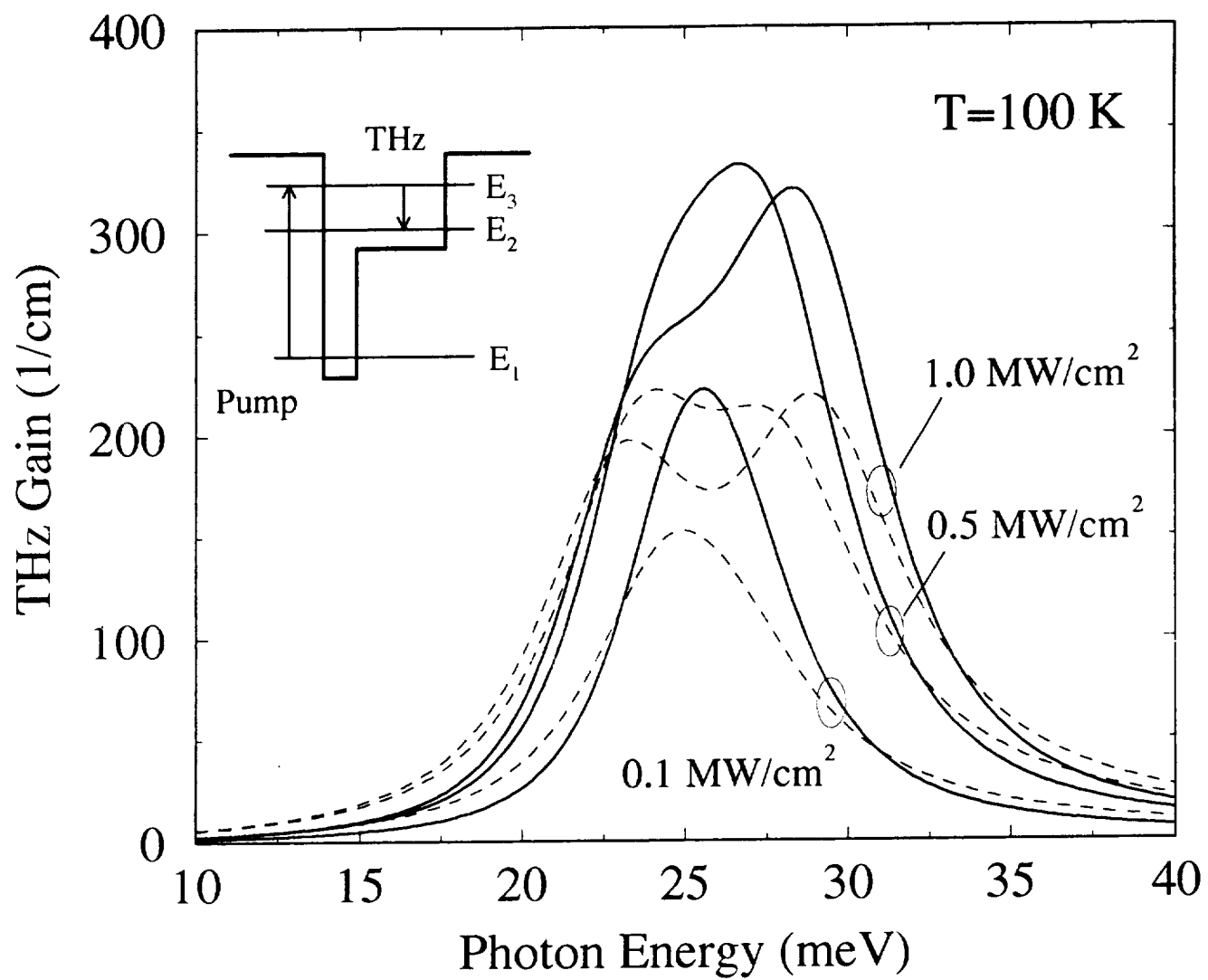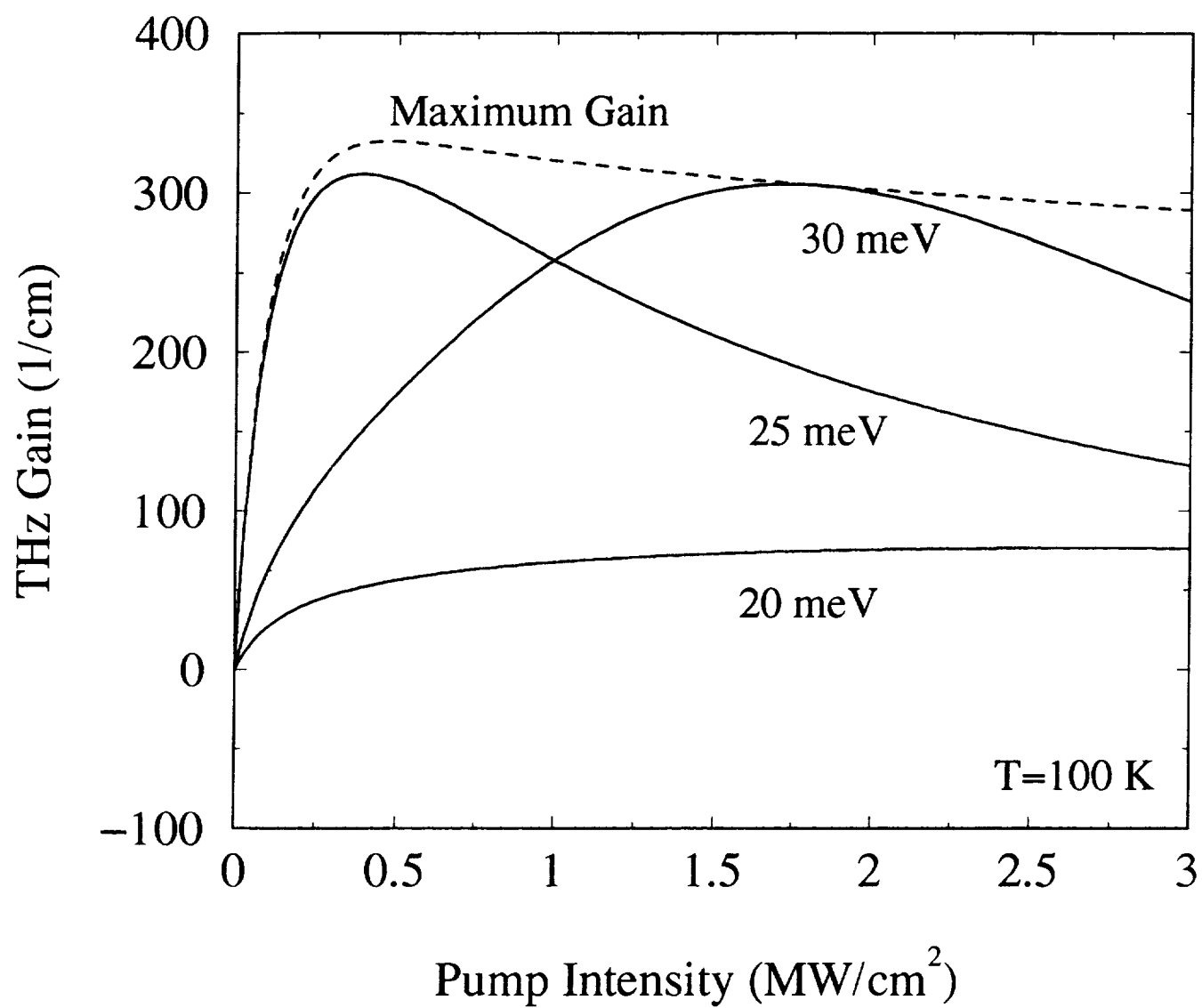FIG. 1

FIG. 2